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13. ABSTRACT (Maximum 200 Words)

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Feasibility of Plasma Aftertreatment for Simultaneous Control of NO_x and Particulates

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ABSTRACT

Plasma reactors can be operated as a particulate trap or as a NO_x converter. Particulate trapping in a plasma reactor can be accomplished by electrostatic precipitation. The soluble organic fraction of the trapped particulates can be utilized for the hydrocarbon-enhanced oxidation of NO to NO₂. The NO₂ can then be used to non-thermally oxidize the carbon fraction of the particulates. The oxidation of the carbon fraction by NO₂ can lead to reduction of NO_x or backconversion of NO₂ to NO. This paper examines the hydrocarbon and electrical energy density requirements in a plasma for maximum NO_x conversion in both heavy-duty and light-duty diesel engine exhaust. The energy density required for complete oxidation of hydrocarbons is also examined and shown to be much greater than that required for maximum NO_x conversion. The reaction of NO₂ with carbon is shown to lead mainly to backconversion of NO₂ to NO. These results suggest that the combination of the plasma with a catalyst will be required to reduce the NO_x and oxidize the hydrocarbons. The plasma reactor can be operated occasionally in the arc mode to thermally oxidize the carbon fraction of the particulates.

1. INTRODUCTION

There are many types of plasma reactors [1-2]. Electrical discharge reactors using the corona, dielectric-barrier or pellet bed are among the types most commonly investigated for aftertreatment of NO_x and/or particulates in diesel exhaust [3]. These plasma reactors can be operated as a particulate trap [4-6] or as a NO_x converter [7-10].

Particulate trapping in a plasma reactor can be accomplished by electrostatic precipitation. Corona-type reactors operating as electrostatic precipitators for diesel soot are reported in Refs. [4-5]. A ferroelectric pellet bed reactor, also operating as an electrostatic precipitator of diesel soot, is reported in Ref. [6]. Plasma-based traps need periodic regeneration just like any other particulate trap. Regeneration by the plasma can be achieved in a thermal mode or a non-thermal mode.

The plasma reactor can be operated occasionally in the arc discharge mode to thermally oxidize the particulates.

In Ref. [5], this mode is referred to as the self-cleaning phase of the electrostatic muffler. Thermal oxidation of the particulates precipitated on the electrode surface is achieved in very localized regions of high temperature near the tip of the arc microdischarges.

The plasma reactor can also be operated in a non-thermal corona discharge mode to provide continuous oxidation of the particulates. The non-thermal oxidation is presumably accomplished by the O and OH radicals resulting from electron-impact dissociation of oxygen and water vapor molecules, or by negative hydronium cluster ions, [(H₂O)_nO₂⁻], resulting from electron attachment. Ref. [4] observed a self-cleaning effect in the corona mode that correlates with the production of CO and CO₂. Ref. [11] reports on the oxidation of soot in a dielectric-barrier discharge. It was conjectured that OH radicals oxidized the soot; the soot reduction did not occur in the presence of large amounts of CO, presumably because the OH radicals were consumed by CO. As sometimes the case in the non-thermal plasma literature, the power input is reported but the electrical energy density input to the plasma is not analyzed. An analysis of the residence time, power input and discharge volume reported in Ref. [11] indicates that the electrical energy density in the plasma was an unacceptably large value of over 5000 J/L. Each 10 J/L corresponds to roughly 1% of the engine power output. The electrical energy density is the important control parameter that determines the number of free radicals that are produced in a non-thermal plasma [12-14].

NO_x conversion in a plasma is mainly oxidative in diesel engine exhaust because of the high concentration of oxygen [7]. The O radicals oxidize NO to NO₂. In the presence of water vapor, the OH radicals further oxidize the NO₂ to nitric acid. The efficiency for conversion of NO to NO₂ is low at typical engine exhaust temperatures because the O radical also reduces NO₂ back to NO. Hydrocarbons greatly enhance the oxidation of NO to NO₂ and minimize the formation of acids [8]. As a NO_x converter, the plasma requires hydrocarbons to achieve high oxidation efficiency with low electrical energy consumption.

Diesel particulates are composed mainly of the carbon fraction and the soluble organic fraction (SOF). The SOF

could possibly be utilized for the oxidation of NO to NO₂ in a plasma. The NO₂ can then be used to oxidize the carbon fraction, similar to that in a Continuously Regenerated Trap (CRT) [15]. The use of NO₂ for the oxidation of trapped particulates in CRT devices is fairly well established. In CRT, a precious metal catalyst is used to oxidize NO to NO₂ upstream of a particulate filter. The CRT method requires low sulfur fuel because the catalytic oxidation of NO to NO₂ also leads to the oxidation of SO₂ to SO₃. The use of a plasma for the oxidation of NO to NO₂ can make the process more tolerant to the sulfur content of the fuel [16].

The presence of hydrocarbons is critical to the selective partial oxidation of NO to NO₂ in a plasma [8, 16]. These hydrocarbons may be derived from the SOF or added to the exhaust. The concentration of NO in the exhaust determines the amount of hydrocarbon and electrical energy density that has to be provided to the plasma in order to achieve maximum NO_x conversion efficiency. Determining these requirements is important whether the NO₂ is used to enhance lean-NO_x SCR or oxidize the carbon fraction of trapped particulates.

The hydrocarbon oxidation chemistry and the NO oxidation chemistry in the plasma are strongly coupled. However, the electrical energy density requirement for maximum oxidation of NO to NO₂ is much less than that required for complete oxidation of the hydrocarbon to CO_x and H₂O. Partial oxidation products, particularly aldehydes, would be formed in the plasma. These partially-oxidized hydrocarbons may not be detected if one does not monitor the high electrical energy density being delivered to the plasma. Knowing the energy density required for complete oxidation of the hydrocarbon is important whether the hydrocarbon is derived from the SOF of the particulates or added to the exhaust.

There are two possible routes in the oxidation of carbon by NO₂. One case leads to the reduction of NO₂ to N₂; the other case leads to the backconversion of NO₂ to NO. It is known in tests of CRT particulate traps that even though NO₂ is effective in oxidizing particulate, the reduction of NO_x to N₂ is rather limited. In plasma-assisted catalytic reduction of NO_x, the NO₂ plays an important role in enhancing the NO_x reduction efficiency [8]. The fate of NO₂ during the oxidation of carbon is important if one contemplates the use of plasma techniques for the simultaneous removal of NO_x and particulates.

This paper examines the hydrocarbon and electrical energy density requirements in a plasma for maximum NO_x conversion in both heavy-duty and light-duty diesel engine exhaust. The energy density required for complete oxidation of hydrocarbons is also examined and compared to that required for NO_x conversion. The fate of NO₂ in reaction with carbon is investigated.

II. NO_x CONVERSION

Both chemical kinetics calculations and experimental measurements are presented in this section to examine the hydrocarbon and electrical energy density

requirements in the plasma. The plasma chemistry model is the same as that discussed in Penetrante et al.[7, 16]. We used propene as the hydrocarbon surrogate because the chemical reaction database for propene is more established and facilitates comparison of the modeling to experiments. The plasma reactor used in the experiments is a pulsed corona discharge reactor consisting of a metal wire inside a metal cylinder. The plasma chemistry is not peculiar to this type of plasma processor; all electrical discharge plasma reactors accomplish essentially the same gas-phase plasma chemistry under the same gas conditions [12-13]. The important control parameter in the plasma reactor is the electrical energy density delivered to the plasma [12-14].

Figure 1 shows the chemical kinetics calculation of the plasma energy density required to oxidize NO to NO₂ in a gas mixture simulating a heavy-duty diesel engine exhaust with an initial NO concentration of 600 ppm. The oxidation efficiency is shown for different values of C₁/NO_x, where the hydrocarbon additive is propene. In the absence of hydrocarbons, the oxidation efficiency is very low even at high values of electrical energy density input to the plasma. The maximum oxidation efficiency increases as the C₁/NO_x ratio is increased. A C₁/NO_x ratio of 6 is required to get 80% oxidation efficiency. The maximum oxidation efficiency increases slowly as the C₁/NO_x is increased further. At a C₁/NO_x of 6, the energy density required to achieve maximum oxidation efficiency is around 30 J/L. The energy density required for maximum oxidation approaches 20 J/L at the limit of high C₁/NO_x.

Figure 2 shows the chemical kinetics calculation of the plasma energy density required to oxidize NO to NO₂ in a gas mixture simulating a light-duty diesel engine exhaust with an initial NO concentration of 100 ppm. Note that for the same C₁/NO_x ratio, the electrical energy density required by the plasma to achieve the same oxidation efficiency is much less compared to that for the case of a heavy-duty diesel engine. When the initial NO concentration is low, the plasma needs to produce a smaller number of radicals; thus, the plasma requires less electrical energy density. At a C₁/NO_x of 6, the energy density required to achieve maximum oxidation efficiency is only around 6 J/L.

Figure 3 shows the comparison between the model predictions and experimental measurements of the oxidation efficiency, as a function of the electrical energy density input to the plasma, for different initial concentrations of NO, with the C₁/NO_x set at 6. The experiments validate the chemical kinetics calculations fairly well.

Figure 4 shows the comparison between the model predictions and experimental measurements of the electrical energy density required by the plasma, as a function of initial NO concentration, to achieve maximum oxidation of NO to NO₂, when C₁/NO_x is 6. The experiments validate the chemical kinetics calculations over the range of initial NO concentrations relevant to both light-duty and heavy-duty diesel engines.

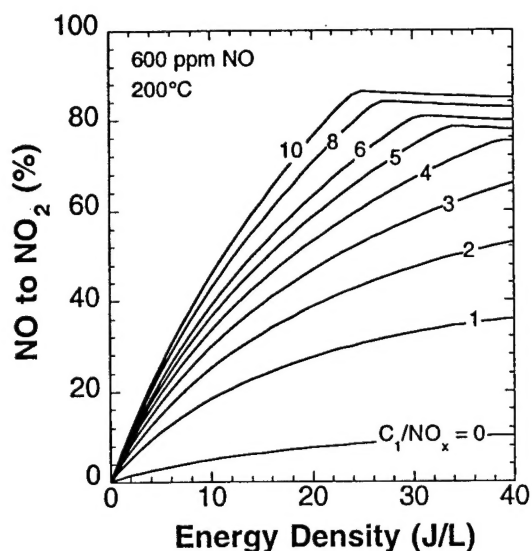


Figure 1. Chemical kinetics modeling of the plasma oxidation of NO to NO₂ in a gas mixture simulating a heavy-duty diesel engine exhaust, using propene additive. Initial NO concentration = 600 ppm, in 10% O₂, 10% CO₂, 5% H₂O, balance N₂. Gas temperature = 200°C.

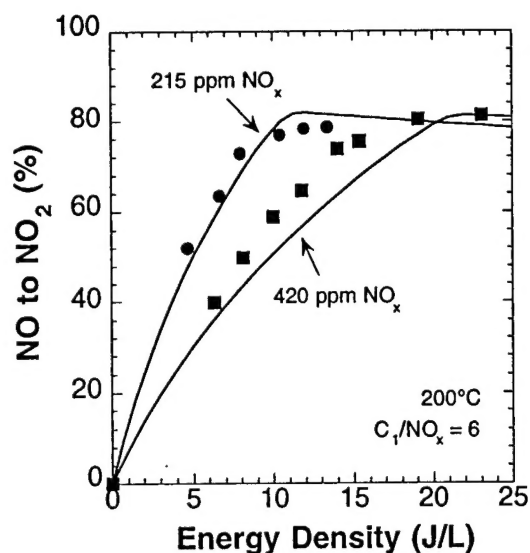


Figure 3. Experimental measurements (points) and modeling predictions (lines) of the plasma oxidation of NO to NO₂ in a gas mixture containing various initial concentrations of NO in 10% O₂, 10% CO₂, 5% H₂O, balance N₂. Propene additive. Gas temperature = 200°C.

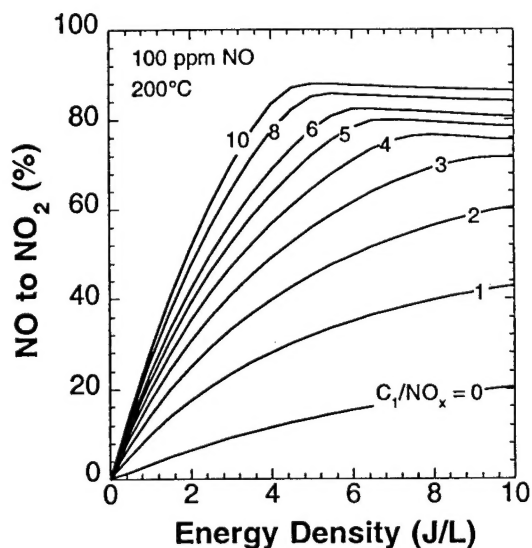


Figure 2. Chemical kinetics modeling of the plasma oxidation of NO to NO₂ in a gas mixture simulating a light-duty diesel engine exhaust, using propene additive. Initial NO concentration = 100 ppm, in 10% O₂, 10% CO₂, 5% H₂O, balance N₂. Gas temperature = 200°C.

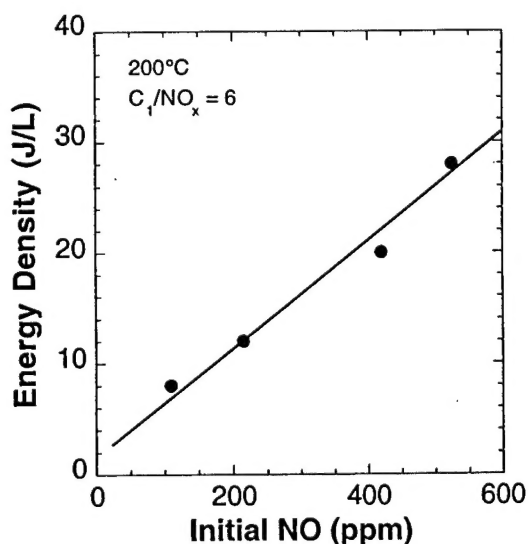


Figure 4. Experimental measurements (points) and modeling prediction (line) of the electrical energy density required by the plasma, as a function of initial NO concentration, to achieve maximum oxidation of NO to NO₂ with C₁/NO_x = 6. Simulated diesel exhaust with 10% O₂, 10% CO₂, 5% H₂O, balance N₂. Propene additive. Gas temperature = 200°C. Line: modeling. Points: experiment.

III. HYDROCARBON OXIDATION

Figure 5 shows the calculated concentration of aldehydes formed during plasma processing of 100 ppm NO in a simulated diesel exhaust at 200°C with a C_1/NO_x of 6. Formaldehyde is the major product of the partial oxidation of propene in the plasma. Note that the electrical energy density required to convert NO to NO_2 is much less than that required to fully oxidize the aldehydes. Whereas it is possible to convert 100 ppm of NO to NO_2 with less than 10 J/L, conversion of the resulting 50 ppm of formaldehyde to H_2O and CO_x requires more than 150 J/L. This result shows that the electrical power required by the plasma to completely oxidize the hydrocarbons is unacceptably large even for light-duty diesel engine exhaust.

Some of the minor products shown in Figure 5 include CH_3CHCO and C_2H_3CHO . Species CH_3CHCO is methyl ketene and species C_2H_3CHO is acrolien. Acrolien is experimentally observed as an intermediate product in the low temperature oxidation of propene [17]. The addition of O atom to propene to form a biradical and its decomposition forms methyl ketene. The rate of this process is based on the current understanding of the O + propene reaction as reviewed in Ref. [18]. Reactions to consume acrolien and methyl ketene by radical attack are included in the detailed chemical kinetic mechanism.

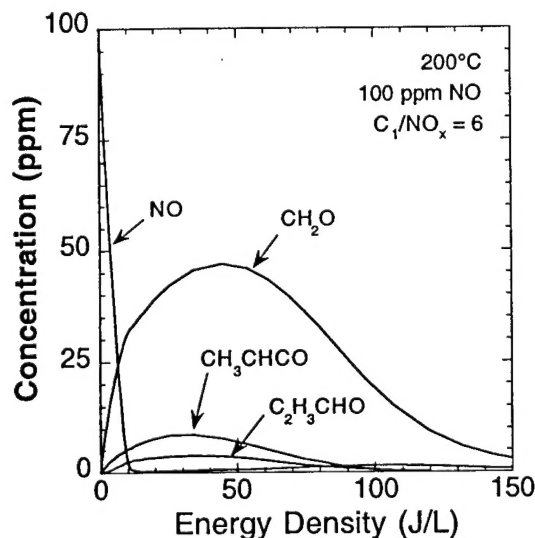


Figure 5. Chemical kinetics calculation of the concentration of aldehydes formed during plasma processing of 100 ppm NO in a simulated diesel exhaust with 10% O_2 , 10% CO_2 , 5% H_2O , balance N_2 . Propene additive, $C_1/NO_x = 6$. Gas temperature = 200°C.

Figure 6 shows the calculated concentration of other species formed during plasma processing of 100 ppm NO in a simulated diesel exhaust at 200°C with a C_1/NO_x of 6. Large amounts of CO are formed during the plasma oxidation of hydrocarbons.

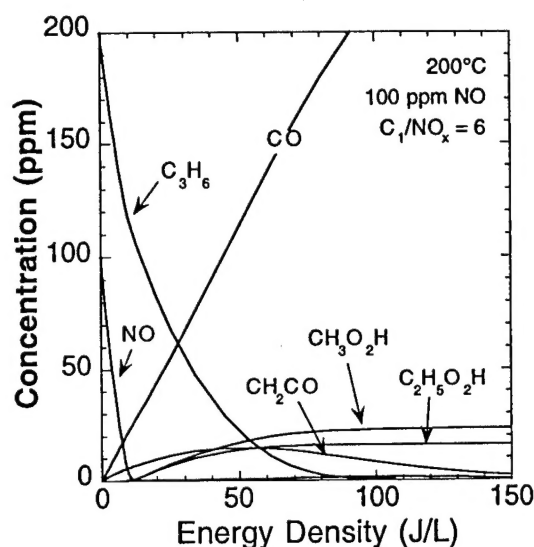


Figure 6. Chemical kinetics calculation of the concentration of various species during plasma processing of 100 ppm NO in a simulated diesel exhaust with 10% O_2 , 10% CO_2 , 5% H_2O , balance N_2 . Propene additive, $C_1/NO_x = 6$. Gas temperature = 200°C.

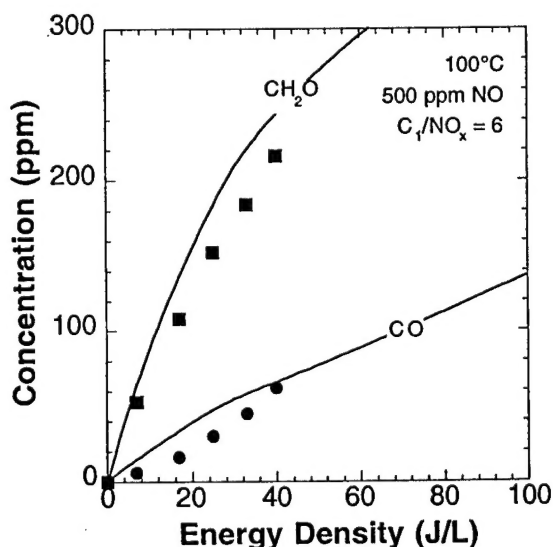


Figure 7. Experimental measurements (points) and modeling predictions (lines) of the concentration of formaldehyde and CO formed during plasma processing of 500 ppm NO in a gas mixture with 10% O_2 , balance N_2 . Propene additive, $C_1/NO_x = 6$. Gas temperature = 100°C.

Figure 7 shows the comparison between the model predictions and experimental measurements of the concentration of formaldehyde and CO formed during plasma processing of 500 ppm NO in a gas mixture with 10% O_2 , balance N_2 , at 100°C with a C_1/NO_x of 6. This level of NO is typical of that in heavy-duty diesel engine exhaust. There is fairly good agreement between the modeling and the experiment. With a C_1/NO_x of 6, about 25 J/L is required to get maximum oxidation of NO to

NO₂. At this energy density, about 150 ppm of formaldehyde has already been formed. The amount of aldehydes increases further as the energy density is increased. Oxidation of these aldehydes will require very large electrical energy density input to the plasma.

IV. CARBON OXIDATION

This section shows the results of experiments on the reaction between NO₂ and carbon pellets in various gas mixtures. The oxidation of the carbon by NO₂ can lead to reduction of NO_x or backconversion of NO₂ to NO. These experiments measure the NO_x reduction efficiency and the amount of backconversion of NO₂ to NO.

Figure 8 shows the fate of NO₂ during its reaction with carbon pellets at 250°C in a gas stream containing only N₂. There is about 20% NO_x reduction. The NO_x remaining after the reaction is composed mostly of NO.

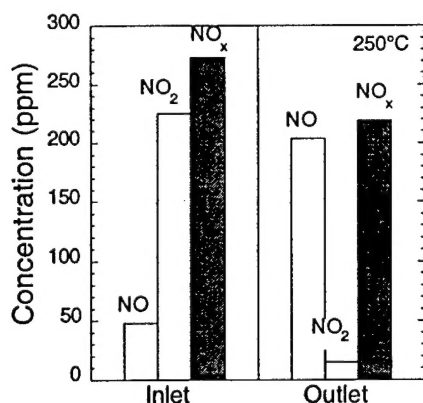


Figure 8. Oxidation of carbon by NO₂ in N₂. Gas temperature = 250°C.

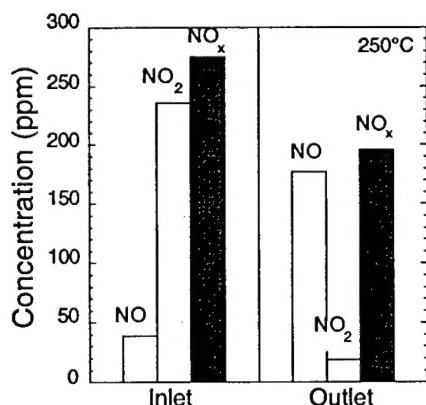


Figure 9. Oxidation of carbon by NO₂ in 10% O₂, balance N₂. Gas temperature = 250°C.

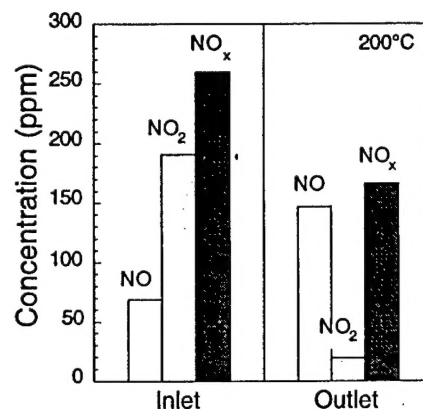


Figure 10. Oxidation of carbon by NO₂ in 10% O₂ + 1500 ppm C₁ kerosene. Gas temperature = 200°C.

Figure 9 shows the same type of experiment conducted in a gas stream containing 10% O₂, balance N₂. The oxygen seems to have promoted the NO_x reduction efficiency to about 30%. Again the NO_x remaining after the reaction is composed mostly of NO.

Figure 10 shows the result of another experiment to simulate the effect of the volatile organic fraction of particulates on the NO₂ reduction. The gas stream in this experiment contains 10% O₂ and 1500 ppm C₁ kerosene. The NO_x reduction efficiency is about 35%, with the remaining NO_x being composed mostly of NO.

The results in Figures 8-10 show that even though the NO₂ could be utilized for the oxidation of the carbon fraction of the particulates, the reaction with the carbon fraction cannot provide a high level of NO_x reduction. Another means will have to be provided to achieve a high NO_x reduction efficiency. If one is going to use plasma-assisted SCR for the NO_x reduction, the regeneration of the particulate trap will have to be done without the NO₂. A way to do this would be to operate the plasma reactor occasionally in the arc mode to thermally oxidize the carbon fraction of the particulates. Ref. [5] describes an example of the effectiveness of this technique and how it can be implemented.

V. CONCLUSIONS

The presence of hydrocarbons is critical to the selective partial oxidation of NO to NO₂ in a plasma. These hydrocarbons may be derived from the soluble organic fraction of the particulates or added to the exhaust. A C₁/NO_x ratio of around 6 is required to achieve optimum oxidation of NO to NO₂ in the plasma. In a light-duty diesel engine exhaust with 100 ppm NO and C₁/NO_x of 6, the electrical energy density required for maximum NO_x conversion is only around 6 J/L. For the same C₁/NO_x, the energy density requirement is linearly proportional to the initial NO concentration. In a heavy-duty diesel engine exhaust with 600 ppm NO, the energy density required goes up to 30 J/L. There is much less penalty for having a plasma in light-duty engine applications.

The electrical energy density required for complete oxidation of hydrocarbons in a plasma is much greater than that required to achieve maximum NO_x conversion. Aldehydes and CO will be formed. The energy density required to oxidize the aldehydes will be unacceptably large even for light-duty applications. A catalyst in combination with the plasma will be required to take care of the aldehydes and CO.

The NO₂ from the plasma can be used to non-thermally oxidize the carbon fraction of trapped particulates. However, the reaction of NO₂ with carbon cannot provide a high level of NO_x reduction and leads mostly to the backconversion of NO₂ to NO. If one is going to use plasma-assisted SCR for the NO_x reduction, the regeneration of the particulate trap will have to be done without the NO₂. A way to do this would be to operate the plasma reactor occasionally in the arc mode to thermally oxidize the carbon fraction of the particulates.

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